

On The Importance of Organic Oxygen for Understanding Organic Aerosol Particles

Y. Pang,¹ B.J. Turpin,² and L.A. Gundel¹

¹Lawrence Berkeley National Laboratory
Environmental Energy Technologies Division
Indoor Environment Department
University of California, Berkeley, CA 94720

²Department of Environmental Sciences
Rutgers University
New Jersey

March 2005

This work has been supported in part by Small Business Technology Transfer Research Phase 1, Award DE-FG02-03ER86168 to Sunset Laboratory, through the US Department of Energy Contract DE-AC03-76SF00098.

ABSTRACT

This study shows how aerosol organic oxygen data could provide new and independent information about organic aerosol mass, aqueous solubility of organic aerosols, formation of secondary organic aerosol (SOA) and the relative contributions of anthropogenic and biogenic sources. For more than two decades atmospheric aerosol organic mass concentration has usually been estimated by multiplying the measured carbon content by an assumed organic mass (OM)-to-organic carbon (OC) factor of 1.4. However, this factor can vary from 1.0 to 2.5 depending on location. This great uncertainty about aerosol organic mass limits our understanding of the influence of organic aerosol on climate, visibility and health.

New examination of organic aerosol speciation data shows that the oxygen content is the key factor responsible for the observed range in the OM-to-OC factor. When organic oxygen content is excluded, the ratio of non-oxygen organic mass to carbon mass varies very little across different environments (1.12 to 1.14). The non-oxygen-OM-to-non-oxygen OC factor for all studied sites (urban and non-urban) is 1.13 ± 0.02 . The uncertainty becomes an order of magnitude smaller than the uncertainty in the best current estimates of organic mass to organic carbon ratios (1.6 ± 0.2 for urban and 2.1 ± 0.2 for non-urban areas). When aerosol organic oxygen data become available, organic aerosol mass can be quite accurately estimated using just OC and organic oxygen (OO) without the need to know whether the aerosol is fresh or aged. In addition, aerosol organic oxygen data will aid prediction of water solubility since compounds with OO-to-OC higher than 0.4 have water solubilities higher than 1 g per 100 g water.

INTRODUCTION

The organic constituents of airborne particles influence the behavior and impacts of particulate matter (PM) on human health, regional visibility and global climate (Novakov and Penner 1993; NRC 1999; IPCC 2001). Organic PM consists of hundreds of compounds from many chemical classes that differ widely in polarity and aqueous solubility. Improved understanding the roles played by aerosol organics in atmospheric processing depends on knowledge of their aggregate polarity and aqueous solubility. One approach is detailed chemical characterization through identification and quantification of individual constituents by gas or liquid chromatography coupled with mass spectrometry (GC-MS and LC-MS, respectively).

The determination of organic compounds by GC-MS or LC-MS generally involves high volume sampling, complex sample preparation (e.g., solvent extraction, evaporation and derivatization), sample analysis and data reduction. The complexity and limits of detection of these methods currently limit their widespread use, although they have been used to identify extractable organic compounds in organic aerosols at various locations (Alves et al., 2002; Edney et al., 2003; Kubatova et al., 2002; Mochida et al., 2003; Rogge et al., 1993; Zheng et al., 2002). However, these approaches have typically identified less than 30% of total organic mass in ambient aerosols. Even though molecular-level analyses are extremely valuable, simpler thermal methods (Ellis et al., 1984; Chow et al., 1993; Birch and Cary, 1996) are more widely used to characterize the carbonaceous components of PM by determining organic and elemental (black) carbon (OC and EC, respectively). Such bulk analyses could provide sufficient data to characterize spatial and temporal distributions of organic PM and estimate organic PM

mass concentrations, if the relationship between OC and organic mass can be established accurately.

However, because of the complexity of organic aerosols, obtaining accurate estimates of organic PM mass concentrations from OC remains a challenge. For 25 years aerosol organic mass estimates have been obtained by multiplying measured OC mass by an assumed OM (organic mass)-to-OC ratio of 1.4 (Turpin and Lim, 2001). This ratio represents an average organic molecular weight per carbon ratio that was calculated more than two decades ago (White and Roberts, 1977) from organic speciation data measured on two days in Pasadena, CA in 1970s. Values of OM-to-OC factor from 0.9 to 2.5 have been reported in the literature (Putaud et al., 2000; Russell, 2003; Turpin and Lim, 2001) although ratios less than 1.0 are not physically meaningful. Turpin and Lim (2001) predicted that the OM-to-OC ratio will see increasing use in the current decade as a practical tool for the development of control strategies for fine particles, even though this method has large uncertainties. Based upon review and analysis of current speciation data for organic aerosols, they recommended ratios of 1.6 ± 0.2 for urban fresh and 2.1 ± 0.2 for non-urban aged aerosols. As an alternative approach, Russell (2003) computed the OM-to-OC ratio from infrared spectroscopic measurements of organic functional groups. This approach requires the functional group standards of absorptivity of the mix of organic compounds in ambient PM (for example, alkanes, aromatics, carbonyls, alcohols and organic acids). Overlapping peaks also complicate quantitation.

Recent reviews (Jacobson et al., 2000; Seinfeld and Pankow, 2003; Huebert and Charlson, 2000) show that the past 30 years of investigation of organic aerosol concentrations and properties have relied heavily on organic carbon measurement.

Aerosol organic oxygen has received little attention, although oxygen, by mass, is second only to carbon in organic PM. The primary objective of this investigation is to use currently available data for aerosol organic composition to evaluate the importance of determination of organic oxygen, for accurate estimation of organic PM mass and prediction of organic aerosol solubility and density. The approach is to start with the framework of Turpin and Lim (2001) and show how uncertainties in estimation of organic mass change when both organic carbon and organic oxygen data are available. The long range objective of this investigation is to provide motivation and justification for updating tools for aerosol organic oxygen so that it can be determined as routinely as OC is at present.

DATA ANALYSIS

OM-to-OC and non-oxygen-OM-to-OC ratios

Below we show that the amount of oxygen is the main factor causing the large variations in OM-to-OC ratios among samples from different sources and locations (Putaud et al., 2000; Russell, 2003; Turpin and Lim, 2001). Since the possible range of C to H molar ratios in organic compounds is quite narrow (1 to 1.4), and aerosol organic nitrogen is expected to be much less prevalent than OO, excluding oxygen from the calculation of aerosol OM-to-OC ratios should also narrow possible range of values. Therefore, what we define as the non-oxygen OM to OC ratio should have lower uncertainty than the OM-to-OC ratio in atmospheric aerosols.

The second column of Table 1 shows the OM-to-OC ratios for compound classes which have been found in ambient aerosols (Turpin and Lim 2001) and the ratios of non-oxygen-OM-to-OC for the same classes.

Table 1. Organic mass to organic carbon ratios, with and without oxygen, for classes of particle-phase organic compounds found in urban air (Rogge et al., 1993; Schauer, 1998).

Compound class	OM-to-OC ^a	Non-oxygen OM-to-OC ^b
n-Alkanes (C ₂₃ –C ₃₄)	1.2	1.2
n-Alkanoic acids (C ₉ –C ₃₂)	1.3–1.5	1.2
n-Alkenoic acids (C ₁₇ –C ₁₈)	1.3–1.5	1.2
n-Alkanols (C ₂₅ –C ₃₂)	1.2	1.2
n-Alkanals (C ₉ , C ₂₆ –C ₃₂)	1.2–1.3	1.2
Aliphatic dicarboxylic acids (C ₂ –C ₉)	1.7–3.8	1.1-1.2
Diterpenoid acids (C ₂₀)	1.3	1.1
Other multifunctional aliphatic acids (C ₃ –C ₆)	2.5–3.1	1.1-1.2
Aromatic polycarboxylic acids (C ₈ –C ₁₀)	1.7–2.1	1.1
Polycyclic aromatic hydrocarbons(C ₁₆ –C ₂₄)	1.0–1.1	1.0-1.1
Polycyclic aromatic ketones and quinones (C ₁₇ –C ₁₉)	1.1–1.2	1.0-1.1
Steroids (Cholesterol, C ₂₇)	1.2	1.2
Nitrogen-containing compounds (C ₆ –C ₁₀)	1.2–1.9	1.2-1.3
Phenol and substituted phenols (C ₆ –C ₇)	1.5	1.1
Guaiacol and substituted guaiacols (C ₇ –C ₁₁)	1.4–1.6	1.1
Syringol and substituted syringols (C ₈ –C ₁₂)	1.4–1.7	1.1
Mono-, sesqui-, and triterpenoids (C ₁₀ –C ₁₂ , C ₃₀)	1.3	1.1-1.2
Sugars (levoglucosan, C ₆)	2.3	1.1
oligomers ^c	1.5-2.1	1.1

^aFrom Turpin and Lim, 2001.

^bThis work.

^cFrom Fig. 3 of Kalberer et al., 2004, assuming three routes for polymer formation.

Indeed, the non-oxygen OM-to-OC ratios have a much narrower range (1.0 to 1.3) than OM-to-OC ratios (1.0 to 3.8). The highest OM-to-OC ratios are obtained for acids, for which the amount of oxygen is the main contributor to the variation of OM-to-OC ratios. For example, the OM-to-OC ratios of oxalic acid ($C_2H_2O_4$) and nonanedioic acid ($C_9H_{16}O_4$) are 3.8 and 1.7, respectively. When oxygen is excluded, the non-oxygen-OM-to-OC ratios of oxalic acid and nonanedioic acid are very similar (1.1 and 1.2, respectively). A new study indicates that oligomers may contribute significantly to organic aerosols (Kalberer et al., 2004). Table 1 shows that non-oxygen-OM-to-OC ratios are very close (1.1) for the oligomers suggested by Kalberer et al.

Table 2 shows what happens to non-oxygen OM-to-OC ratios when the data in Table 1 are used with available concentration data for aerosol organic compounds in urban and non-urban California (Rogge et al., 1993; Schauer, 1998). The ratio of non-oxygen OM-to-OC is 1.13 ± 0.02 for measured organic compounds for all sites (urban and non-urban).

Table 2. Average OM-to-OC and non-oxygen-OM-to-OC ratios calculated as mass-weighted averages, based on ambient organic speciation data and Table 1.

Location	OM-to-OC ratio	non-Oxygen-OM -to- OC	% mass ^a	Reference
Los Angeles Basin (Annual avg.)			7-15%	Rogge et al., 1993
Downtown	1.65	1.11		
West LA	1.63	1.12		
Rubidoux	1.77	1.11		
San Nicolas Island ^b	1.29	1.14		
San Joaquin Valley (Dec 26, 1995; Jan 4, 1996)				Schauer, 1998
Fresno	2.54 ^c ; 2.24 ^d	1.14 ^c 1.14 ^d	5–10%	
Bakersfield	2.52; 2.58	1.14;1.14	5–10%	
Kern Refuge	1.82; 3.04	1.14;1.14	<1%	

^a % mass is the percentage of the organic mass identified at the molecular level and used in the ratio.

^b Numerous compounds were below detection limits at this background site.

^c for Dec 26 1995

^d for Jan 4, 1996

Large uncertainties exist in the OM-to-OC molecular weight ratio for organic aerosol PM because of the lack of information about polar organic PM components. Turpin and Lim (2001) suggested that current analytical limitations result in underestimation of the polar organic constituents in PM and OM-to-OC ratios, that are based on molecular level analysis. Because polar organics have more oxygen than non-

polar organics, analytical limitations have more influence on OM-to-OC than non-oxygen-OM-to-OC ratios.

Solubility of organic aerosols

The aqueous solubility of the complex mixture of organic species in PM can influence chemical and physical transformation of atmospheric aerosols. For example, modeling results suggest that water soluble organics in aerosols influence the number and size of droplets by increasing the droplets' solute mass, and reducing both the surface tension and the critical supersaturation ratio for droplet activation (Anttila and Kerminen, 2002). Water soluble organics have more significant effects on climate than insoluble organics (Swietlicki et al., 1999; Anttila and Kerminen, 2002; Chung and Seinfeld, 2002; Shantz et al., 2003;). After surveying the literature, Saxena and Hildemann (1996) concluded that 20-70% of the organic aerosol mass consists of water-soluble organics. Tables 3 and 4 show OM-to-OC, non-oxygen OM-to-OC and Oxygen-to-C ratios for organic compounds with low and high aqueous solubilities, respectively, for species included in the review by Saxena and Hildemann (1996). We did not include aromatic polycarboxylic acids in Tables 3 and 4 because their water solubility varies from 0 to 3 g solute per 100 g water, bridging both categories.

Table 3. Mass to carbon ratios for organic compounds with low aqueous solubility (<1 g per 100 g water) that have been found in ambient aerosols.

Compound class	OM-to-OC	non-Oxygen- OM-to-OC	Oxygen-to- Carbon
n-Alkanes	1.2	1.2	0
n-Alkanoic acids (C ₉ –C ₃₀)	1.3–1.5	1.2	0.1-0.3
Diterpenoid acids (C ₂₀)	1.3	1.1	0.1
Polycyclic aromatic hydrocarbons	1.1	1.1	0
Polycyclic aromatic ketones and quinones	1.1-1.2	1.0-1.1	0.1

Table 4. Mass to carbon ratios for organic compounds with substantial aqueous solubility (>1 g per 100 g water) that have been found in ambient aerosols.

Compound class	OM-to-OC	non-Oxygen- OM-to-OC	Oxygen-to-Carbon
Aliphatic dicarboxylic acids (C ₂ –C ₆)	1.7–3.8	1.1-1.2	0.9-1.8
Glyoxal (C ₂)	2.4	1.1	1.3
Ketoacids (C ₂ –C ₅)	1.9–3.1	1.1	1.0-2.0
Polyols (C ₂ –C ₇)	1.5-2.6	1.1	0.4-1.3
p-Nitrophenol (C ₆)	1.9	1.3	0.7
Amines and amino acids	2.4–3.6	1.4-1.9	0.0-1.3 (0.4-1.3 without amines)
Misc. multifunctional (C ₃ –C ₆)	2.4–3.1	1.1-1.2	1.3-2.0

Table 3 shows that most of the less soluble organic compounds have oxygen-to-carbon-mass ratios from 0.0 to 0.3. However, Table 4 shows that the oxygen-to-carbon ratios for water-soluble organics are larger, 0.4- 2.0, except for amines that comprise a very small part of the total aerosol organic mass.

Organic aerosol density

Table 5 shows the densities of aerosol organic compounds, as measured or estimated for 20 °C and 1 atm (from Howard and Neal, 1992; Immirzi and Perini, 1977; Lang, 1973; Lide, 1994, as identified by Rogge et al., 1993 or Sempere and Kawamura, 1994). When the mass ratios of oxygen-to-carbon are higher than 0.5, the densities are higher than 1.0 g/cm³, except PAHs.

Table 5. Densities of particle-phase organic compounds and oxygen-to-carbon mass ratios.

Compound class	Density Range g cm ⁻³	Density Avg. g cm ⁻³	Oxygen- to-Carbon
n-Alkanes (C ₂₃ –C ₃₄)	0.77–0.81	0.79 ± 0.01	0.0
n-Alkanoic acids (C ₉ –C ₃₀)	0.81–0.99	0.89 ± 0.07	0.1-0.3
n-Alkanals (C ₉)	0.83	0.83	0.2
Aliphatic dicarboxylic acids (C ₂ –C ₉)	1.03–1.90	1.46 ± 0.23	0.6-1.8
Ketocarboxylic acids (C ₂ –C ₃)	1.1–1.27	1.18 ± 0.12	1.0-2.0
Aromatic polycarboxylic acids (C ₈ –C ₁₀)	1.39–1.60	1.49 ± 0.08	0.6-1.1
Polycyclic aromatic hydrocarbons (C ₁₆ –C ₂₄)	1.10–1.55	1.28 ± 0.12	0.0
Polycyclic aromatic ketones and quinones (C ₁₇ , C ₁₈)	1.16–1.22	1.19	0.1
Steroids (Cholesterol, C ₂₇)	1.07	1.07	0.05
Nitrogen-containing compounds (C ₆ –C ₁₀)	1.08–1.19	1.11 ± 0.05	0.0-0.3
Polyols (C ₂ –C ₈)	0.93–1.26	1.04 ± 0.13	0.4-1.3
amino acids (C ₂ –C ₆)	1.01–1.60	1.42 ± 0.21	0.4-1.3

DISCUSSION

Turpin and Lim (2001) found that OM-to-OC ratios vary considerably across the classes of compounds found in PM, and they found large differences between rural and urban locations, presumably because of increased oxidation with regional transport, as well as differences in emission sources. In contrast, Table 2 shows that the non-oxygen-OM-to-OC ratios varied little (from 1.11 to 1.14 with $1.13 \text{ mean} \pm 0.02$). For Kern Refuge, the OM-to-OC ratios were quite different for Dec 1995 and Jan 1996 (1.82 and 3.04 respectively). However, the non-oxygen-OM-to-OC ratios were the same. For San Nicolas Island the OM-to-OC ratio was lower than that expected for a non-urban area, but many samples from San Nicolas Island had organic concentrations below detection limits. In spite of this, the non-oxygen-OM-to-OC ratio at this site was closer to the ratio for non-urban areas (e.g., Fresno, Kern Refuge, 1.14) than for urban areas (e.g., LA Downtown, West LA, 1.11).

The degree of saturation of carbon bonds increases during oxidation of both polar and non-polar organic compounds. Saturated organic compounds generally have more hydrogen bonded to carbon. Furthermore, high molecular weight polar compounds are generally more difficult to identify and yet may account for a large fraction of organic aerosol mass. The unidentified polar organic compounds will certainly influence the true OM-to-OC ratios, but the non-oxygen-OM-to-OC ratios are much less affected. This may explain why the non-oxygen-OM-to-OC ratio of San Nicolas with more aged secondary organic aerosol (SOA) was different from that of downtown LA with more primary organic aerosols even though fewer organic compounds were identified.

The uncertainty in the non-oxygen-OM-to-OC ratio (1.13 ± 0.02) is an order of magnitude smaller than the uncertainty in the best current estimates of total organic mass to the organic carbon ratios (1.6 ± 0.2 for urban and 2.1 ± 0.2 for non-urban areas). Therefore, when aerosol organic oxygen concentrations can be determined directly from ambient samples, and if their sampling and measurement uncertainties are reasonable ($\sim 15\%$ or less) the sum of aerosol organic oxygen mass and non-oxygen organic mass calculated from OC data will provide much more accurate estimates of aerosol organic mass than are now possible. Compared with determination of all organic compounds or functional groups in aerosols, determination of aerosol organic oxygen mass is likely to be easier and more cost-efficient.

The data of Table 1 as discussed above indicate that oxygen is the main factor contributing to the variation of OM-to-OC ratios among compound classes that have been quantified in ambient PM. Table 2 shows that variation in organic oxygen is largely responsible for the range of OM-to-OC ratios found in ambient aerosols. Therefore, monitoring aerosol organic oxygen concentrations may provide useful information for source apportionment, investigations of aerosol processing, and prediction of effects.

Identification of the relative contribution of anthropogenic and biogenic sources is critical for policy makers. From data for fireplace combustion of pine, oak, and eucalyptus wood (Schauer et al., 1998) we calculate that the ratios of concentration-weighted aerosol organic oxygen mass to carbon mass were 0.77, 0.81, and 0.96, respectively, and averaged 0.85. This is much higher than we calculate from the data of Schauer et al. (2002) for fossil fuel combustion for which emissions were dominated by hydrocarbons (the concentration weighted oxygen-to-carbon ratio was 0.1 for catalyst-

equipped gasoline-powered motor vehicle engine(s)). This example suggests that the ratio of aerosol organic oxygen mass to organic carbon mass might also provide insights concerning the relative contribution of biogenic and anthropogenic sources to organic aerosols.

The degree of oxidation, inferred from the amount of organic oxygen in aerosol organics, is also an indicator of SOA formation because SOA consists of the oxidized products of reactions of primary organic compounds with photochemically generated oxidants (e.g., ozone and OH). Three methods are used currently to estimate SOA concentrations in the atmosphere (Seinfeld and Pankow, 2003): (a) the EC tracer method, (b) Detailed speciation and quantification of members of organic compound classes; and (c) Determination of concentrations of molecular tracers to assess the role of individual hydrocarbon precursors in SOA formation. Seinfeld and Pankow (2003) concluded that the EC tracer method will continue in common use because better methods are currently lacking, even though the estimates of SOA formation from OC and EC data have large uncertainties. Measurements of aerosol organic oxygen (OO), used with OC and EC data, might reduce EC tracer method uncertainties because increases in OO are likely to track atmospheric oxidation processes. OO measurements can provide a new tool to monitor formation of SOA and subsequent atmospheric reactions. Adding OO characterization to OC-EC measurements could also improve source apportionment of ambient PM. OO along with OC-EC data may help indistinguishing oxygen-rich primary biogenic aerosol from SOA produced from both anthropogenic and biogenic sources.

To our knowledge, no methods can predict or estimate the solubility of aggregated organic aerosols. Table 3 and Table 4 suggest that aerosol organic oxygen

could aid the predication of organic aerosol water solubility when OC-EC data are also available. Such data could contribute to improved prediction of the effects of organic aerosol on climate, visibility and dosimetry.

In conclusion, aerosol organic oxygen data could provide new and independent information about organic aerosols and new tools to understand several properties of organic aerosols. Such oxygen data could provide a better estimation of aerosol organic mass and aqueous solubility, as well as better apportionment of the contributions of anthropogenic, biogenic sources and secondary organic aerosol formation. We recommend the use of a non-oxygen-organic mass to organic carbon ratio 1.13 ± 0.02 for organic aerosol mass estimation and development of a direct measurement of aerosol organic oxygen. This measurement approach will decrease uncertainties in organic PM mass and improve our understanding of effects of organic aerosols on health, visibility, and climate. Benner and Hansen (1984) developed a thermal method for aerosol oxygen and demonstrated its usefulness for characterization of urban PM (Benner et al., 1984). Although their method has not been widely used, efforts have begun to update and optimize it for routine measurements of aerosol organic oxygen (Cary et al. 2003).

ACKNOWLEDGMENTS

The authors gratefully acknowledge Tica Novakov for inspiring discussions and Elizabeth Stewart for technical assistance. This work has been supported in part by Small Business Technology Transfer Research Phase 1 Award DE-FG02-03ER86168 to Sunset Laboratory via a subcontract to E.O. Lawrence Berkeley National Laboratory, through the US Department of Energy Contract DE-AC03-76SF00098.

REFERENCES

- Alves, C., A. Carvalho and C. Pio (2002). Mass balance of organic carbon fractions in atmospheric aerosols, *J. Geophys. Res.-Atmospheres*, 107 (D21), 8345.
- Anttila, T., and V.M. Kerminen (2002). Influence of organic compounds on the cloud droplet activation: A model investigation considering the volatility, water solubility, and surface activity of organic matter, *J. Geophys. Res.-Atmospheres*, 107 (D22), 4662.
- Benner, W.H., and A.D.A. Hansen (1984). Temperature-programmed evolved gas analysis for determining pyrolyzable oxygen, *Anal. Chem.* 56: 2876-2879.
- Benner, W.H., A.D.A. Hansen, L. Gundel, and T. Novakov (1984). Determination of organic oxygen in ambient particulate matter, *Sci. Total Environ.* 36: 271-276.
- Birch, M.E. and RA Cary (1996). Elemental carbon-based method for monitoring occupational exposures to particulate diesel exhaust, *Aerosol Sci. Technol.* 25: 221-241.
- Cary, R.A. Gundel, L. A. and Pang, Y. (2003) Advance real-time monitoring of particulate carbon, oxygen and mass, Small Business Technology Transfer Research Grant No. DE-FG02-03ER86168, U.S. Department of Energy.
- Chow J.C., J.G. Watson, L.C. Pritchett, W.R. Pierson, C.A (1993). Frazier and R.G. Purcell, The DRI thermal optical reflectance carbon analysis system - description, evaluation and applications in the United States air quality studies, *Atmos. Environ.-A.* 27: 1185-120.
- Chung, S.H., and J.H. Seinfeld (2002) Global distribution and climate forcing of carbonaceous aerosols, *J. Geophys. Res.-Atmospheres*, 107 (D19), 4407.
- Dean, J.A., ed. (1973), *Lange's Handbook of Chemistry, 11th ed.*, McGraw-Hill, New York, 1600 pp.
- Edney, E.O., T.E. Kleindienst, T.S. Conver, C.D. McIver, E.W. Corse, and W.S. Weathers (2003). Polar organic oxygenates in PM_{2.5} at a southeastern site in the United States, *Atmos. Environ.* 37: 3947-3965.

- Ellis E.C., T.Novakov, M.D.Zeldinn (1984) Thermal characterizat on of ofganic aerosols, *Sci. of the Total Environ.* 36: 261-270.
- Grosjean, D., and S.K Friedlander (1975). Gas-particle distribution factors for organic and other pollutants in the Los Angeles atmosphere, *J. Air Pollut. Contr. Assoc.* 25: 1038-1044.
- Howard, P.H., and M. Neal (1992). *Dictionary of Chemical Names and Synonyms*, Lewis, New York, 2527 pp.
- Huebert, B.J., and R.J. Charlson (2000) Uncertainties in data on organic aerosols, *Tellus Series B-Chem. & Phys. Meteorology*, 52: 1249-1255.
- Immirzi, A., and B. Perini (1977) Prediction of density in organic crystals,, *Acta Crystallogr. Sect. A*, 33: 216-218.
- IPCC (2001). *Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment*, Report of the Intergovernmental Panel on Climate Change. Cambridge, United Kingdom and New York, NY, USA, Cambridge University Press.
- Jacobson M.C., H.C. Hansson. K.J.Noone and R.J. Charlson (2000). Organic atmospheric aerosols: Review and state of the science. *Rev. Geophys.*, 38: 267-294.
- Japar, S.M., A.C. Szkarlat,, R.A. Gorse, Jr., E.K. Heyerdahl, R.L. Johnson, J.A. Rau, J.J .and Huntzicker (1984). Comparison of solvent extraction and thermal optical carbon analysis methods: application to diesel vehicle exhaust aerosol, *Environ. Sci. Technol.* 18: 231-234.
- Kubatova, A., R. Vermeylen, M. Claeys, J. Cafmeyer, and W. Maenhaut (2002). Organic compounds in urban aerosols from Gent, Belgium: Characterization, sources, and seasonal differences - *J. Geophys. Res.-Atmospheres*, 107 (D21), 8343.
- Lide, D.R. (1994). *Handbook of Chemistry and Physics*, CRC Press, Ann Arber, MI.
- Mochida, M., K. Kawamura, N. Umemoto, M. Kobayashi, S. Matsunaga, H.J. Lim, B.J. Turpin, T.S. Bates, and B.R.T. Simoneit (2003). Spatial distributions of oxygenated organic compounds (dicarboxylic acids, fatty acids, and levoglucosan) in marine aerosols over the western Pacific and

off the coast of East Asia: Continental outflow of organic aerosols during the ACE-Asia campaign, *J. of Geophys. Res.-Atmospheres*, 108 (D23), 8638.

National Research Council. *Research Priorities for Airborne Particulate Matter; II. Evaluating Research Progress and Updating the Portfolio*; National Academy Press: Washington, DC, 1999; p 27-38.

Novakov, T. and J.E Penner (1993). Large contribution of organic aerosol to cloud-condensation-nuclei concentrations, *Nature* 365: 823-826.

Putaud, J.P., R. Van Dingenen, M. Mangoni, A. Virkkula, F. Raes, H. Maring, J.M. Prospero, E. Swietlicki, O.H. Berg, R. Hillamo, and T. Makela (2000). Chemical mass closure and assessment of the origin of the submicron aerosol in the marine boundary layer and the free troposphere at Tenerife during ACE-2, *Tellus B-Chem. Phys. Meteorol*, 52: 141-168.

Rogge, W.F., M.A. Mazurek, L.M. Hildemann, G.R. Cass, and B.R.T. Simoneit (1993). Quantification of urban organic aerosols at a molecular level - identification, abundance and seasonal variation, *Atmos. Environ.* 27: 1309-1330.

Russell, L.M. (2003). Aerosol organic-mass-to-organic-carbon ratio measurements, *Environ. Sci. Technol.* 37: 2982-2987.

Saxena, P., and L.M. Hildemann (1996), Water-soluble organics in atmospheric particles - a critical review of the literature and application of thermodynamics to identify candidate compounds, *J. Atmos. Chem.* 24: 57-109.

Schauer, J.J. (1998). Source Contributions to Atmospheric Organic Compound Concentrations: Emissions Measurements and Model Predictions, Ph.D. Thesis., Ph.D. California Institute of Technology, Pasadena, CA.

Schauer, J.J., M.J. Kleeman, G.R. Cass, and B.R.T. Simoneit (2002). Measurement of emissions from air pollution sources. 5. C-1-C-32 organic compounds from gasoline-powered motor vehicles, *Environ. Sci. Technol.* 36: 1169-1180.

Seinfeld, J.H., and J.F. Pankow (2003). Organic atmospheric particulate material, *Ann. Rev. Phys. Chem.* 54: 121-140.

Sempere, R., and K. Kawamura (1994). Comparative distributions of dicarboxylic acids and related polar compounds in snow rain and aerosols from urban atmosphere, *Atmos. Environ.* 28: 449-459.

Shantz, N.C., W.R. Leaitch, and P.F. Caffrey (2003). Effect of organics of low solubility on the growth rate of cloud droplets - art. no. 4168, *J. Geophys. Res.-Atmos.* 108 (D5), 4168.

Swietlicki, E., J.C. Zhou, O.H. Berg, B.G. Martinsson, G. Frank, S.I. Cederfelt, U. Dusek, A. Berner, W. Birmili, A. Wiedensohler, B. Yuskiewicz, and K.N. Bower (1999). A closure study of sub-micrometer aerosol particle hygroscopic behaviour, *Atmos. Res.* 50: 205-240.

Turpin, B.J., and H.J. Lim (2001). Species contributions to PM_{2.5} mass concentrations: Revisiting common assumptions for estimating organic mass, *Aerosol Sci. Technol.* 35: 602-610.

White, W.H., and P.T. Roberts (1977). On the nature and origins of visibility reducing aerosols in the Los Angeles Air Basin, *Atmos. Environ.* 11: 803-812.

Zheng, M., G.R. Cass, J.J. Schauer, and E.S. Edgerton (2002). Source apportionment of PM_{2.5} in the southeastern United States using solvent-extractable organic compounds as tracers, *Environ. Sci. Technol.* 36: 2361-2371.